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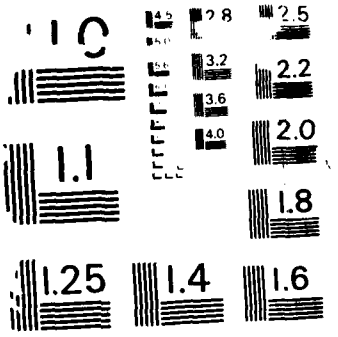

THE EFFECTS OF THIN COMPRESSIVE FILMS ON INDENTATION FRACTURE TOUGHNESS MEASUREMENTS

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Abstract

Thin compressive films have been shown to decrease the lengths of radial cracks produced by a Vickers indentation in a variety of nonmetallic materials. The intrinsic stress of submicron thick films deposited by reactive ion beam sputtering was measured by a cantilever technique. The change in the apparent indentation fracture toughness of the coated material was correlated with film thickness and stress, indentation load, and the nature of the substrate.

INTRODUCTION

The indentation fracture technique is a simple and rapid means of determining the fracture toughness, K_c, of brittle materials. In this method, a Vickers-shaped diamond indenter of a given load is used to produce radial cracks that can be viewed on the sample surface. The lengths of the cracks can then be directly related to the fracture toughness.[1]

A thin stressed layer on the surface of a brittle material can significantly modify the lengths of the radial cracks and therefore also the computed value of the toughness. This phenomenon has been observed in ion-implanted materials by several groups.[2-4] Lawn and Fuller[5] have developed a model that relates thin-layer surface stresses to changes in indentation crack dimensions. Their model predicts a change in the computed fracture toughness, K, that is independent of indenter load. However, work with ion implantation surface layers[3,4] shows that the change in K is smaller at larger loads than at smaller loads.

In this paper, a systematic study is described in which changes in the indentation fracture toughness of a variety of brittle materials are correlated with thin film stress and thickness, and with indenter load.

EXPERIMENTAL

Reactive ion beam deposition was used to deposit thin (< 1 μm) Si_3N_4, SiO_2 and Al_2O_3 films from elemental and ceramic targets. Depositions were made at room temperature. These sputtered films are intrinsically stressed as depicted in Fig. 1. This figure shows a model of the relaxed stress distribution of a film/substrate system.[6] The compressive stress in the film is balanced by a tensile stress in the bulk which will cause an unclamped substrate to bend. The resultant stress is highest near the film-substrate interface. This stress is intrinsic to the growth conditions and does not develop as a result of a

mismatch in the thermal expansion coefficients of the film and substrate. Furthermore, for amorphous or polycrystalline films it is expected to be independent of the nature of the substrate.

The intrinsic stresses of the films used in this study were measured by a sensitive bending plate technique with capacitive detection.[7] For this technique, a thin glass cover slip is coated on one side with 20 nm of aluminum metal to make it conductive. The cover slip is then clamped at one end so that it forms a parallel plate capacitor with a metal plate. When a stressed film is deposited on the uncoated side of the cover slip, the glass bends and the resulting change in capacitance is used to measure the film stress. Typically, 10 - 50 nanometers of film were deposited; the ion gun was turned off and the capacitive system was allowed to equilibrate thermally before the stress was determined. Additional depositions were made until 500 nm had been deposited.

Corroboration of the cantilever determined stress valves was obtained by measuring the stress of a 400 nm thick Si_3N_4 film on a thin, round ZnS substrate. Film stress caused the ZnS substrate to assume a spherical curvature that was measured by counting the Newton's rings under a sodium lamp. The seven rings that were measured from a 0.4 micron thick Si_3N_4 film corresponded to a film stress of 1.4 GPa. This is reasonably close to the 1.6 GPa value obtained by the cantilever system. The cover-slip cantilever system is coated stationary, while all of the other substrates in this study were mounted on a rotating planetary substrate holder that produced uniform film coverage.

The Vickers indent technique was used to measure the fracture toughness. The fracture toughness, K_c, in an unstressed material is given by[1]

$$K_c = \chi P/c_o^{3/2} \quad (1)$$

where P is the load, and c_o is the radial crack length defined in Fig. 2. The coefficient χ is defined by $\chi = (0.016)(E/H)^{1/2}$, where E and H are the Young's modulus and hardness, respectively. This technique provides a measure of the fracture toughness that is independent of the indentor load. Figure 3a shows a glass sample that has been indented with a 1.0 kg load. The glass slide was broken along two of the cracks to expose the radial cracks lying beneath the surface. The full penny-like (or semicircular) shape of the radial cracks did not develop because of a lateral crack 40 μm below the surface. Note how the radial cracks intersect the surface at right angles.

A film-stressed system can alter the size and shape of the penny-like radial cracks as well as the indenter loading required to produce four symmetrical cracks. This crack alteration produces a different apparent fracture toughness. Figure 3b shows the effect of adding a 0.4 μm thick Si_3N_4 film. The radial cracks are deflected near the surface which results in a shorter crack length on the surface. The shapes of the cracks are modified at a depth beneath the surface that is many times the film thickness. Deflected radial cracks were also observed for films on single crystal silicon substrates.

Films up to 1 μm thick were initially grown on 1.2 mm thick soda lime glass (Erie Scientific, New Hampshire) and 0.3 mm thick (100) single crystal silicon substrates. Indents were made at several loadings and crack lengths were measured at least one day later. The delay between indentation and crack analysis was established since cracks in glass may continue to grow after the

load is removed due to moisture assistance. The rate of this postindentation growth decreases with time.[1] Silicon does not exhibit postindentation growth. Measurements made immediately after indentation provide a more accurate K_c for unstressed materials.[1] The indenter was aligned so that the cracks in silicon were always oriented along the <100> directions. K was calculated for indents with four radial cracks and with $c \geq 2b$. This put a lower limit on the loads and the depths that were sampled. The hardness of the samples, which was also obtained from the Vickers indent, was not noticeably changed by the addition of these films.

RESULTS AND DISCUSSION

The cantilever stress system gives values of σ_s equal to 0.2, 1.0 and 1.6 GPa for films of Al_2O_3, SiO_2 and Si_3N_4, respectively. In all cases the accumulated stress was found to increase linearly with thickness.

While the stress of the SiO_2 films is less than that of the Si_3N_4, it was observed that on glass the threshold load for crack production was higher with the SiO_2 films. Glass with a 1.0 μm thick SiO_2 film required greater than a 1.5 Kg load for any radial cracks to form while uncoated glass gave four cracks with a 0.2 Kg load. Glass with a 0.5 μm thick Si_3N_4 film requires a 0.4-0.7 Kg load to crack. Thus, with an SiO_2 film the crack initiation load increased by an order of magnitude. Greater adhesion or better protection against moisture-assisted crack growth in the SiO_2/glass system than in the Si_3N_4/glass system may be responsible for the larger threshold load.

As a first approximation to the change in fracture toughness, Lawn and Fuller[5] have derived a thin-film stress intensity factor for a penny-like crack system

$$K_f = 2\gamma\sigma_s d^{1/2} \tag{2}$$

where d is the film thickness, γ is a crack geometry term taken to be unity, and σ_s is the average surface stress of the film. Implicit in the derivation of Eq. (2) is the assumption that the crack remains penny-like, which is not the case, as is shown in Fig. 3. Lawn and Fuller pointed out that this non penny-like shape will lead to some uncertainty in Eq. (2).

The measured apparent fracture toughness, K, is given by the sum of the uncoated fracture toughness plus the thin film stress intensity factor:

$$K = \chi P/c^{3/2} = K_c + K_f = K_c + 2\sigma_s d^{1/2} \quad . \tag{3}$$

Using the cantilever stress measurements, calculated K_f values for 1.0 μm thick films are 3.2, 2.0 and 0.4 $MPam^{1/2}$ for Si_3N_4, SiO_2 and Al_2O_3, respectively. Equation (3) predicts an increased fracture toughness which is independent of load and sampling depth. Equation (3) also predicts that the magnitude of the increased fracture toughness will be independent of the substrate since σ_s in the K_f term is independent of the substrate. The effect of tensile stress in the substrate may reduce the fracture toughness below that predicted by Eq. (3) and this decrease may be dependent on load.

Figure 4 shows the indent results for an uncoated glass and a Si_3N_4-coated glass sample. The measured fracture toughness of the coated glass is a strong function of load and therefore of sampled depth. In addition, the minimum load at which c = 2b is higher for the coated sample than for the uncoated sample. As was mentioned earlier, with coated glass and with some of the other substrates tested (except silicon), the threshold load required to produce any radial cracks is increased considerably. This indicates that the coatings inhibit crack initiation.

The surface fracture toughness was taken as the value of K at a fixed crack length, c', from a least squares fit of K vs $c^{1/2}$. Values of c' were chosen to be as small as possible for each substrate in order to maximize the measured change in fracture toughness. The values of c' for glass and silicon substrates were 60 μm and 16 μm, respectively. Figures 5 and 6 show these surface fracture toughness values as a function of the square-root of the film thickness.

Several other substrates were tested with Si_3N_4 and/or Al_2O_3 coatings. Only a few samples were coated in these cases so the data should be considered less reliable. Table 1 gives the uncoated fracture toughness, the crack length at which the surface toughness was determined, and the increased fracture toughness extrapolated to a 1.0 μm thick film using Eq. (2) (i.e., as $d^{1/2}$). Also listed are the average grain sizes of the materials.

In most cases, the measured K_f is 2 to 6 times smaller than that predicted by Eq. (3). This is most likely caused by the presence of the tensile stress in the substrate or by the deviation from the penny-like shapes of the

cracks. Those sampled substrates with the smaller cracks tended to give larger changes. When the silicon and glass data are plotted versus $c^{1/2}$ and extrapolated to zero crack length, the changes in toughness are approximately twice as high as those listed in Table 1.

Very high toughening was found for single crystal sapphire and no change was observed for fine grained Si_3N_4 and ZnS. These anomalous results indicate that the toughening effect as measured by the Vickers indentation method may be dependent upon the mechanisms by which the surface stresses are accommodated in the substrate. The role of grain size and grain boundaries must be considered as must the levels of stresses in the bulk materials.

Complementary to this work, Burnett and Page[8] have made indent fracture measurements on ion-implanted sapphire. The implanted sapphire expands and produces a thin compressive layer near the surface. Using an implantation depth of 0.2 μm for 5.8×10^{17} Y^+ ions/cm^2 at 300 KeV and a stress of 5 GPa, the calculated K_f is 10 times larger than that measured using a 0.2 Kg load.

CONCLUSIONS

Thin compressive films deposited onto brittle substrates are found to increase the measured indentation fracture toughness of a variety of materials. For a given substrate, the change in fracture toughness with respect to film stress, film thickness, and indent load can be modeled semi-quantitatively. Anomalously large increases are found for single crystal sapphire while no change is measured with polycrystalline ZnS or Si_3N_4.

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FIGURE CAPTIONS

Fig. 1 Stress distribution in a brittle material with a compressive surface film.

Fig. 2 Schematic representation of a Vickers indent in a coated material.

Fig. 3 Glass samples with a 1.0 kg load indent taken with an optical microscope. A) no film and B) with a 0.4 μm thick Si_3N_4 film. The arrows point to the ends of the surface radial cracks.

Fig. 4 Measured fracture toughness of uncoated glass and glass coated with a 0.7 μm layer of Si_3N_4 as a function of indent load.

Fig. 5 Surface fracture toughness of glass coated with Si_3N_4, SiO_2, and Al_2O_3 plotted as the 1/2 power of film thickness.

Fig. 6 Fracture toughness of single crystal silicon coated with Si_3N_4, SiO_2 and Al_2O_3 plotted as the 1/2 power of the film thickness.

Table 1

Measured Fracture Toughness Data from Coated Ceramic and Single Crystal Substrates.

Substrate	Grain Size (μm)	K_c ($MPam^{1/2}$)	c' (μm)	Increase in Toughness K_f ($MPam^{1/2}$) 1 μm Coating		
				Si_3N_4	SiO_2	Al_2O_3
ZnS	4	0.8	100	<0.1		<0.1
Si_3N_4	0.5	4.1	100	< 0.1		
Glass	Amorphous	0.62	60	0.5	0.3	0.17
Glass	Amorphous	0.62	60	(ZnS Coated)*		0.2
As_2S_3	Amorphous	0.16	50			0.10
Silicon	Single Xtal	0.74	16	0.7	0.4	0.3
Sapphire**	Single Xtal	1.7	16	1.7		1.5
ALON	75	1.43	16			0.5
Germanium	> 100	0.5	15	0.6		
Spinel	50	1.1	12			0.6
Calculated***				3.2	2.0	0.4

*ZnS film deposited by laser evaporation and independently determined to have an intrinsic stress of 0.2 GPa

**(0001) surface

***Using Eq. (3).

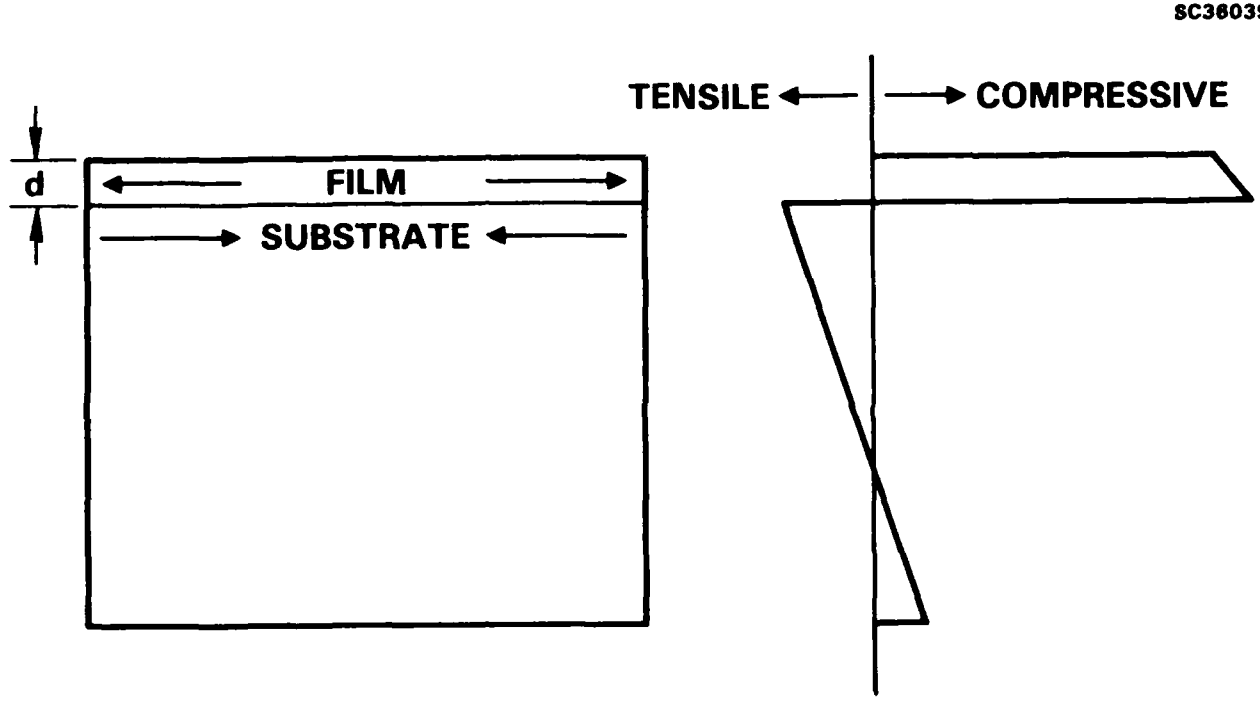

Fig. 1

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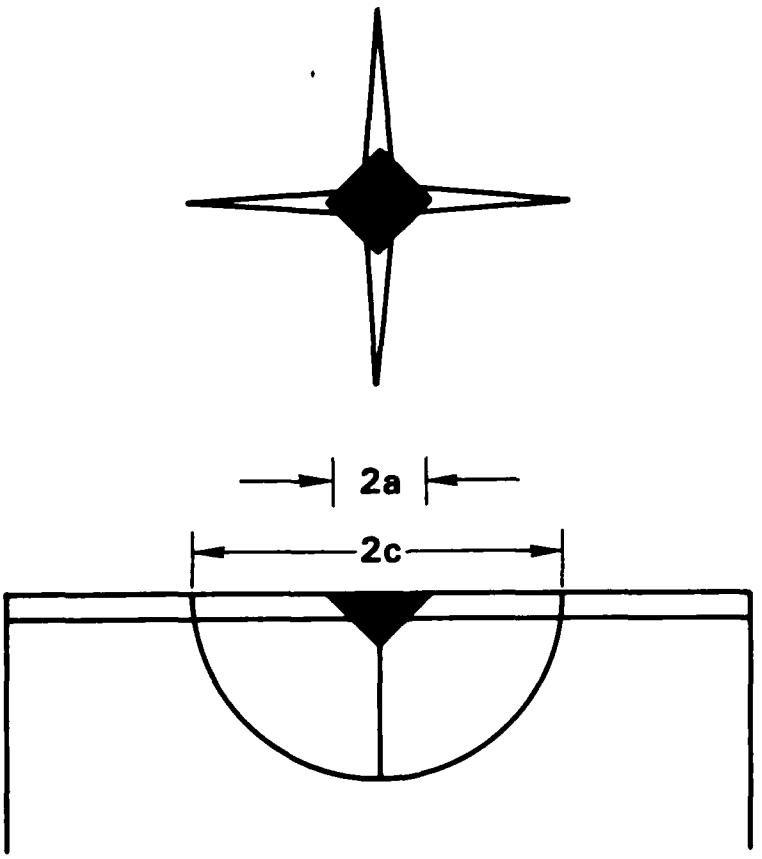

Fig. 2

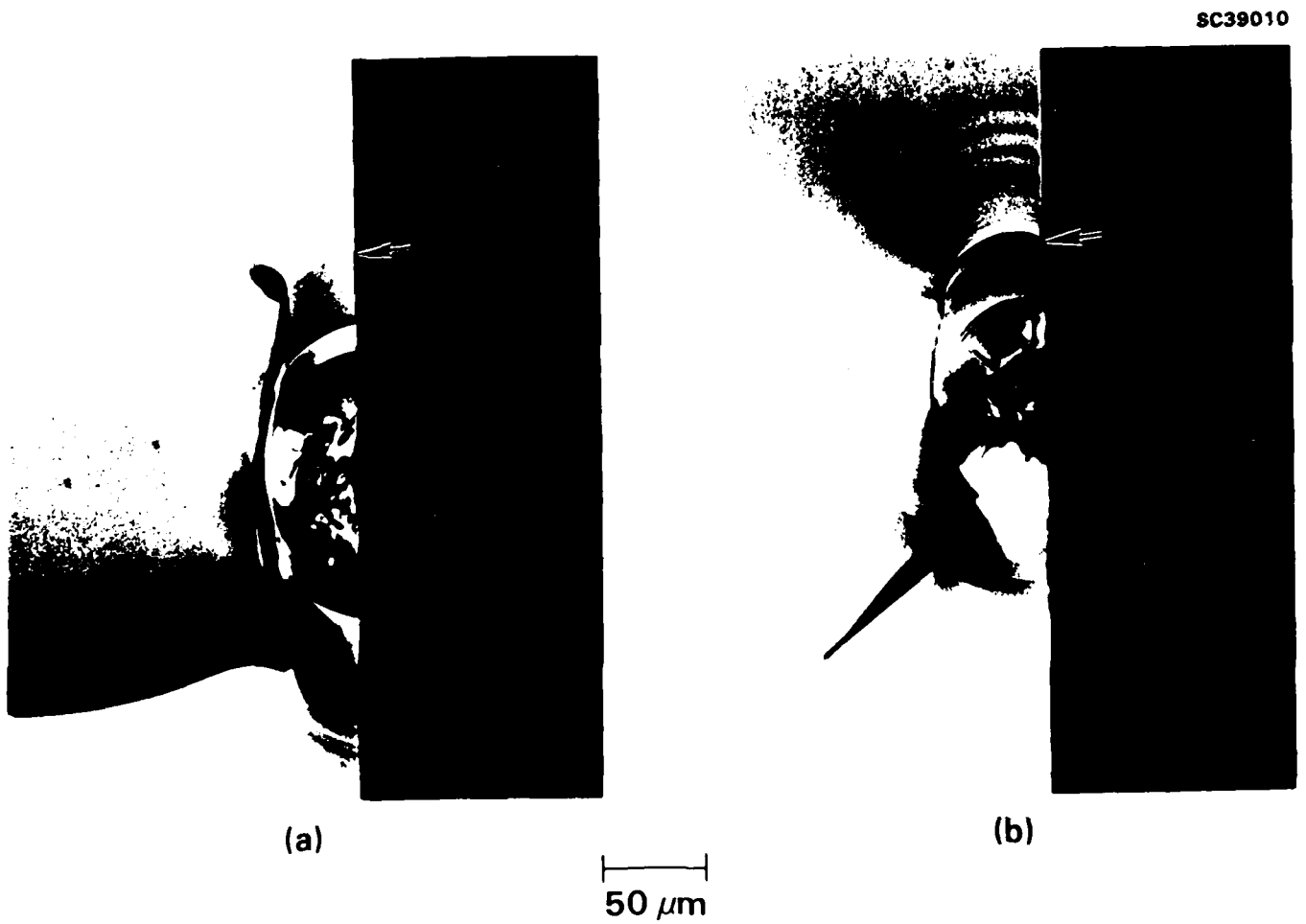

Fig. 3

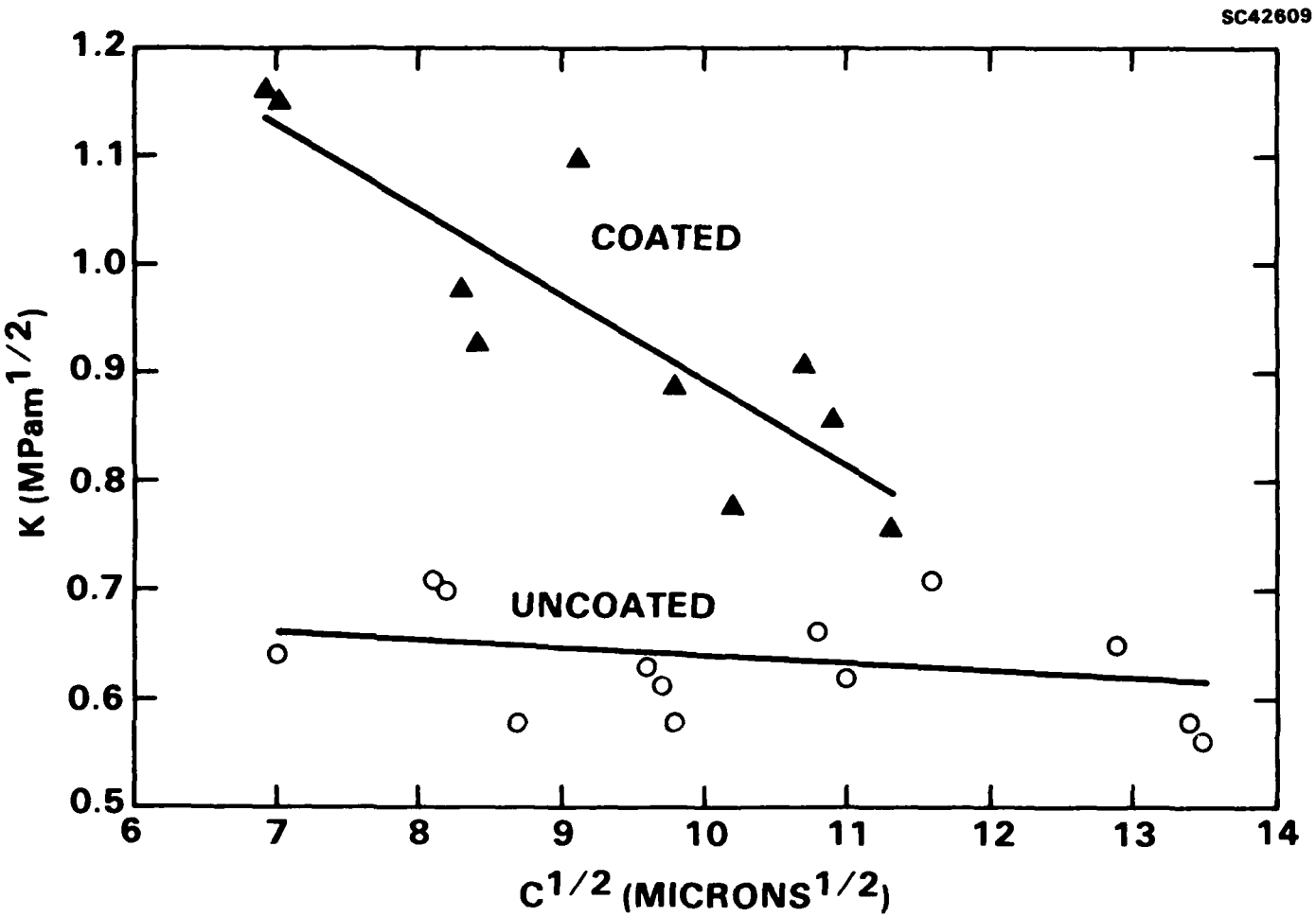
SC42609
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
K (MPam$^{1/2}$)
COATED
UNCOATED
6
7
8
9
10
11
12
13
14
C$^{1/2}$ (MICRONS$^{1/2}$)

Fig. 4

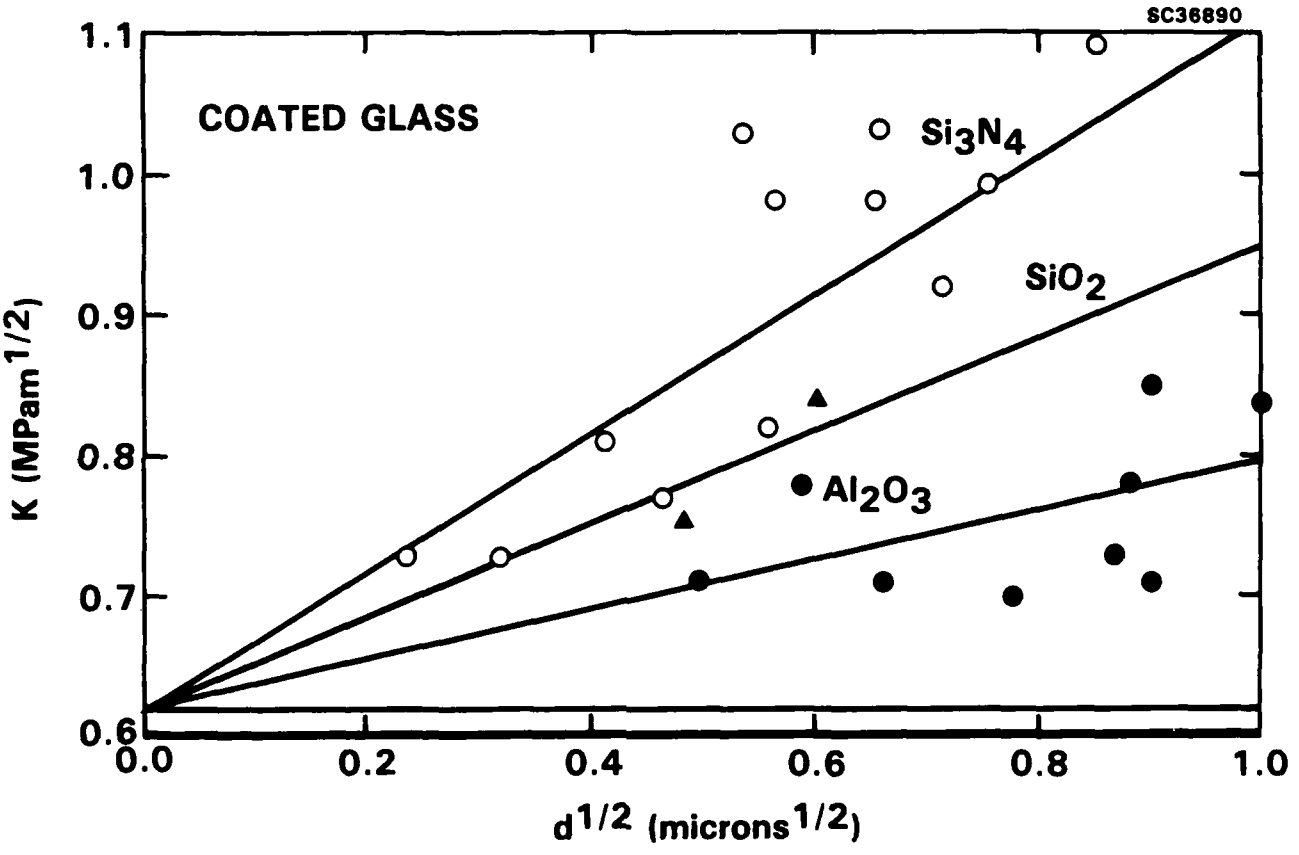
SC36890
COATED GLASS
Si_3N_4
SiO_2
Al_2O_3
K (MPam$^{1/2}$)
d$^{1/2}$ (microns$^{1/2}$)
1.1
1.0
0.9
0.8
0.7
0.6
0.0
0.2
0.4
0.6
0.8
1.0

Fig. 5

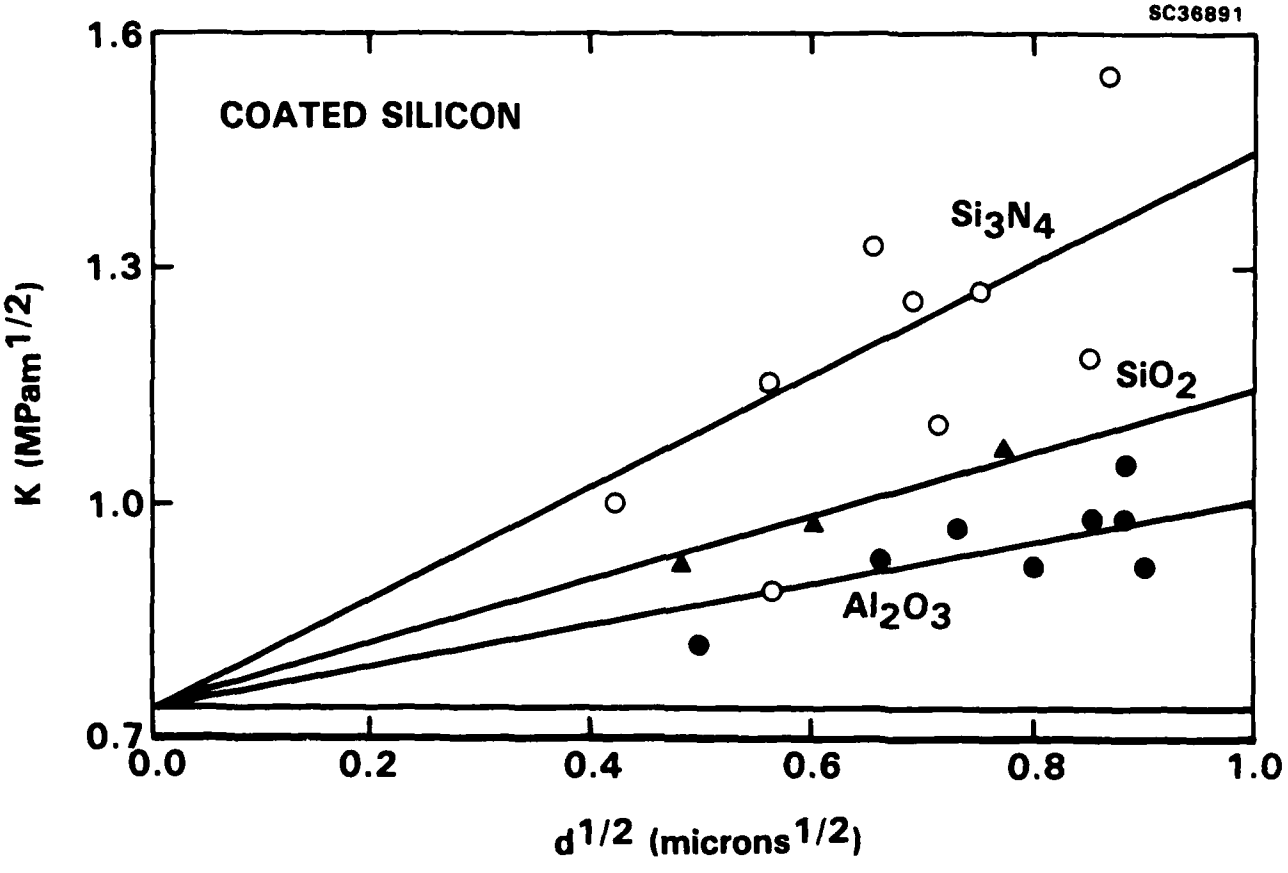
SC36891
COATED SILICON
Si_3N_4
SiO_2
Al_2O_3
K (MPam$^{1/2}$)
1.6
1.3
1.0
0.7
0.0
0.2
0.4
0.6
0.8
1.0
d$^{1/2}$ (microns$^{1/2}$)

Fig. 6

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